

## THE BEHAVIOUR OF HIGHVALE AND VESTA COALS UNDER CO-PROCESSING CONDITIONS

L.K. Lee and B. Ignasiak

Alberta Research Council  
Coal and Hydrocarbon Processing Department  
1 Oil Patch Drive, Bag #1310  
Devon, Alberta  
Canada, T0C 1E0

### INTRODUCTION

Co-processing involves the conversion of relatively low-cost coal and heavy oil in the presence of hydrogen to synthetic crudes that can be further upgraded into premium liquid fuels. This alternative coal conversion concept eliminates or significantly reduces the need for solvent recycle and hence offers greater potential for improving economic performance over current direct coal liquefaction technologies.

Alberta is endowed with large reserves of subbituminous coals, bitumen and heavy oils. The combined processing of these feedstocks is attractive for the Alberta scenario. Alberta Research Council has been active in developing the co-processing technology (1,2). Due to the high oxygen and mineral matter contents in Alberta subbituminous coals plus the high sulfur and metal contents in bitumens, special considerations are necessary when processing these feedstocks, for example, removal of oxygen to avoid excessive hydrogen consumption; removal of mineral matter to reduce deactivation of expensive catalysts; or use of less efficient, low-cost disposable catalysts. Alberta Research Council developed a two-stage process which involves a first-stage coal solubilization in bitumen or heavy oil using a mixture of carbon monoxide and steam in the presence of an alkali metal catalyst, followed by a catalytic hydrogenation second-stage (3).

This paper is focused on the use of disposable iron oxide catalysts in co-processing two Alberta subbituminous coal and bitumen feed pairs. Effects of reaction severity, pressure and coal concentration will be discussed.

### EXPERIMENTAL

#### Process Unit

Co-processing experiments were carried out in a two-stage, continuous flow reactor system with a nominal capacity of 2 Kg/h of slurry feed. The reactor system consists of two stirred tanks of one and two litres in volume connected in series. Products were collected over a material balance period of eight hours under steady state conditions. For all experiments reported in this paper, the material balances were within 100  $\pm$  3%. For comparative purposes, all data were normalized to give 100% recovery by adjusting the weight of the slurry product. Details of the process unit are available elsewhere (4).

Slurry products were distilled according to a modified ASTM D-1160 procedure, followed by extraction using pyridine in a soxhlet apparatus.

#### Feedstocks

The co-processing characteristics of two feed pairs, Highvale (subbituminous B) / Athabasca bitumen and Vesta (subbituminous C) / Cold Lake bitumen, were evaluated. The two coal samples were obtained from the Alberta Research Council's sample bank.

The Athabasca bitumen sample was a Suncor coker feed (IBP 222°C) while the Cold Lake sample was an atmospheric distillation residuum obtained from the Imperial Oil Strathcona Refinery, Edmonton. Analyses of the coals and bitumen samples are given in Tables 1 and 2, respectively.

### Reaction Severity

Previous batch autoclave experiments (5) indicated that iron oxides can be used as effective catalysts to solubilize Alberta coals in petroleum derived solvents at temperatures up to 470°C under a hydrogen atmosphere of about 21 MPa. In order to avoid excessive coking, the continuous flow CSTR experiments were carried out at temperatures below 470°C.

Reaction temperature and space time in both reactors were varied to determine the impact of reaction severity on product yields and qualities. As a first approximation, a simplistic approach was taken by defining a reaction severity index which combined the effects of the temperature (T) and nominal residence time (N. RTime) in a reactor.

$$\text{Reaction Severity (SV)} = [ (N. \text{RTime}) * \text{EXP}(-E/RT) ]_{1\text{st reactor}} \\ + [ (N. \text{RTime}) * \text{EXP}(-E/RT) ]_{2\text{nd reactor}}$$

The activation energy (E) for both stages was assumed to be  $1.26 \times 10^5$  joules/g-mole (30 Kcal/g-mole). A reference severity index of unity was arbitrarily defined at the condition of 400°C for 60 min.

Figure 1 compares co-processing results obtained at various degrees of reaction severity ( $2 < \text{SV} < 5$ ) for the Vesta/Cold Lake pair with moisture-and-ash-free (maf) coal loadings between 27 and 30 wt%. The simplistic, reaction severity index approach gives good correlation with product yields when expressed in terms of maf feed basis. Yields of carbon oxides and hydrogen sulfides are relatively constant over the whole range of conditions studied. Also, as the reaction severity increases the hydrocarbon gas yield increases proportionately, while the pyridine insoluble organic matter (IOM) decreases. Even at the highest reaction severity conditions tested, there was no sign of excessive coke formation, as reflected by the steady decrease in the amount of IOM in the residual solids. These IOM values correspond to coal conversions (measured by pyridine extraction) between 93-99 wt% maf coal, which were found to be in agreement with coal liquefaction results obtained under similar process conditions (7).

With increasing severity distillable oils (IBP-500°C) are formed at the expense of pyridine extractable materials. Using the two-stage CSTR system, a maximum yield of 62 wt% maf feed was obtained at the highest severity tested. Distillable oil yields of as high as 75% maf feed were observed using a tubular reactor system (6).

Comparing product yields from co-processing of the two feed pairs (refer to Figure 1 and 2), the Highvale/Athabasca pair seems to produce more carbon oxides, hydrocarbon gases and IOM, but less distillable oils. These differences which become more pronounced at higher reaction severity conditions, cannot be correlated to the elemental and petrographic analyses as given in Table 1. Although Vesta and Highvale coals have a similar vitrinite content, Vesta coal contains a higher percentage of inertinite and thus is expected to be less reactive. However, conversions of Highvale coal (82-85 wt%) were in fact 10-14 wt% lower than Vesta coal. A similar result was also obtained under the direct coal liquefaction mode (7). On the other hand, the difference in carbon oxides yield from co-processing contradicts previous results obtained for these coals under liquefaction mode (7).

Figures 1 and 2 reveal similar yields of non-distillable, pyridine extractable material for both feed pairs. However, detailed compound class analysis (3) suggests

that most of the residual oils occurred in the form of hydrocarbons, resins and asphaltenes with less than 30 wt% as preasphaltenes. The higher preasphaltene content in products from the Highvale/Athabasca pair further confirmed the lower reactivity of this feed pair.

### Pressure

Effect of system pressure was studied over a narrow range of 14.2 to 20.7 MPa at two different reaction severities. The low pressure experiments were carried out at a slightly lower maf coal loading of 27 wt%, while the high pressure data were collected using 30 wt% of coal. Considering the slight difference in coal loading, there are no significant effects on product distribution due to the lower operating pressure, as indicated in table 3. However, low pressure operating conditions, which may create long term operating problems, warrant further investigation in a large scale pilot unit.

### Coal Concentration

Bitumen is considered a poorer solvent than coal-derived liquids for coal solubilization. In once-through mode operation, the ability of bitumen to solubilize coals may deteriorate as the coal concentration increases. As illustrated in Figure 4, produce yields from co-processing of Highvale/Athabasca under relatively mild severity conditions increase steadily as the coal concentration increases from 0 to 35 wt%. The bitumen-only case gives 58 wt% (feed bitumen basis) of distillable oils. The incremental oil yield at a coal loading of 35 wt% is about 18 wt% on a bitumen feed basis. Translating back to maf coal basis, this is equivalent to 33 wt%, which is 10-15 wt% higher than the typical values when anthracene oil was used as a solvent, under similar process conditions. The higher incremental oil yield provides additional incentive for co-processing of coals and bitumen.

### CONCLUSION

Co-processing of Alberta Subbituminous coals and bitumen in a two-stage, once-through mode is technically feasible. These results indicate that synergistic effects occur when the Highvale and Vesta coals are processed with Athabasca and Cold Lake bitumen, respectively. Decreasing the system pressure from 20.7 to 14.2 MPa has no significant effect on product distribution. Despite the over simplification of the two-stage CSTR model, good correlations are obtained between product yields and the reaction severity index.

### ACKNOWLEDGEMENT

The experimental work reported in this paper was conducted by L. Logan, R. Hawkins, J. Wilson, B. Doherty, and D. Rohatynski. The authors would like to thank Dr. S. Chakrabarty for his comments and reviewing the manuscript. This work was jointly funded by the Alberta Research Council and the Alberta Office of Coal Research and Technology.

### REFERENCES

1. E. Boomer and A. Saddington, *Canadian Journal of Research*; **12**, p825, 1935.
2. B. Ignasiak, L. Lewkowicz, G. Kovacic, T. Ohuchi and M.P. du Plessis, (Editors: S. Kaliaguine and A. Makay); *Catalysis on the Energy Scene*, Elsevier Science Publishers, B.V., Amsterdam, 1984.
3. B. Ignasiak, T. Ohuchi, P. Clark, D. Aitchison, and T. Lee; *ACS Division of Fuel Chemistry*, **31**, No. 4, p200, 1986.

4. L.K. Lee; Alberta Energy and Natural Resource Department / Alberta Research Council (ENR/ARC) Report for Project #34, Final Report for April to September 30, 1985.
5. L. Lewkowicz, R. Lott, S.K. Chakrabartty and B. Ignasiak; ENR-ARC Report, Report # YCLQ-33, 1986.
6. L.K. Lee, D. Aitchison, J. Mendiuk, and G. Kovacic; CCLC Pyrosol Evaluation Program, Phase I Final Report (confidential report), June 1987.
7. L.K. Lee, P.D. Clark, and D.W. Aitchison; ENR-ARC Coal Conversion Research Program, Final Report for 1984/1985, 1, 1986.

Table 1. Typical Properties of Highvale and Vesta Coals

	Highvale	Vesta
ASTM Rank	Subbit. B	Subbit. C
<u>Proximate Analysis (wt%)</u>		
Capacity Moisture	19.6	24.7
Moisture Free Ash	11.4	12.4
Volatile Matter (maf)	39.6	41.9
Fixed Carbon (maf)	60.4	58.1
<u>Elemental Analysis (wt%)</u>		
Carbon	75.0	74.4
Hydrogen	4.3	5.0
Nitrogen	1.1	1.5
Sulfur	0.2	0.6
Oxygen	19.3	18.5
H/C	0.69	0.81
<u>Petrographic Composition (vol%)</u>		
Vitrinite	75.0	73.0
Liptinite	2.1	0.3
Semi-fusinite	22.2	8.9
Inertinite	1.1	17.0

Table 2. Typical Analysis of Feed Oils

	Athabasca Coker Feed	Cold Lake Atm. Bottom
<u>Elemental Analysis (maf)</u>	wt%	wt%
Carbon	82.9	83.2
Hydrogen	10.3	10.3
Nitrogen	0.5	0.9
Sulfur	5.3	5.3
Oxygen (by diff.)	1.1	0.3
H/C	1.49	1.49
<u>Distillation and Extraction</u>		
Moisture	-	0.01
IBP-500°C	29.5	29.2
Residual Oils	69.9	70.7
Residual Solids	0.6	0.05
<u>Compound Class Analysis</u>		
Hydrogen & Resin	79.8	70.5
Asphaltene	18.9	27.2
Preasphaltene	1.3	2.3

Table 3. Effect of Pressure on Co-processing of Vesta/Cold Lake Atmospheric Bottom Using Iron Oxide as Catalysts

	High Severity		Low Severity	
	Case A	Case B	Case C	Case D
Pressure (MPa)	20.7	14.3	20.7	14.2
Temperature (°C)	431/431	431/430	420/419	418/416
Severity Index	3.52	3.90	2.65	2.56
<u>Product Distribution (maf wt% feed)</u>				
C1 - C5	4.53	5.20	3.15	4.08
IBP-200°C	11.14	14.32	7.23	10.43
200-375°C	10.93	16.58	8.54	10.40
375-500°C	31.86	28.76	31.27	29.11
500°C+	36.56	33.81	44.10	42.90
IOM	1.33	1.73	1.76	2.61
H <sub>2</sub> consumed	1.86	2.85	1.65	2.42

Figure 2

Co-processing of Highvale Coal  
and Athabasca Bitumen (Suncor  
Coker Feed)

(using dispersed iron oxide  
as catalyst)

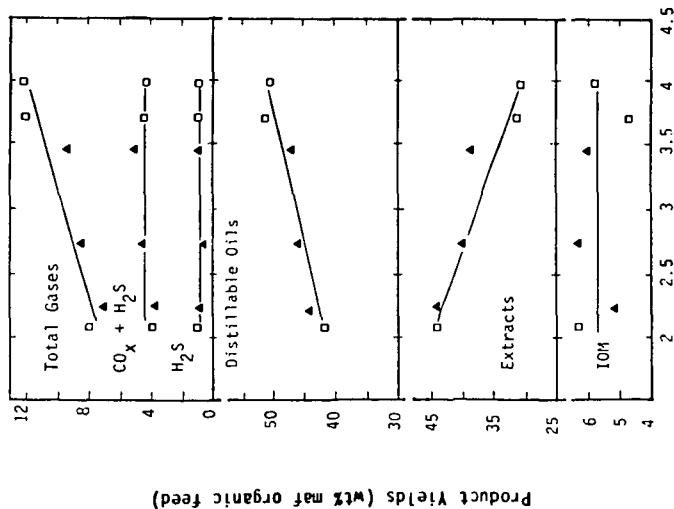


Figure 1

Co-processing of Vesta Coal and  
Cold Lake Atmospheric Bottom

(using dispersed iron oxide  
as catalyst - bench unit)

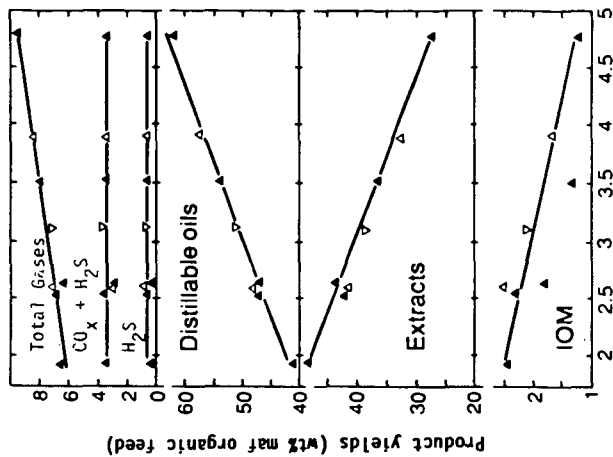


Figure 4

Effects of Coal Concentration  
- low severity (Highvale Coal/  
Athabasca bitumen, iron oxide  
as catalyst)

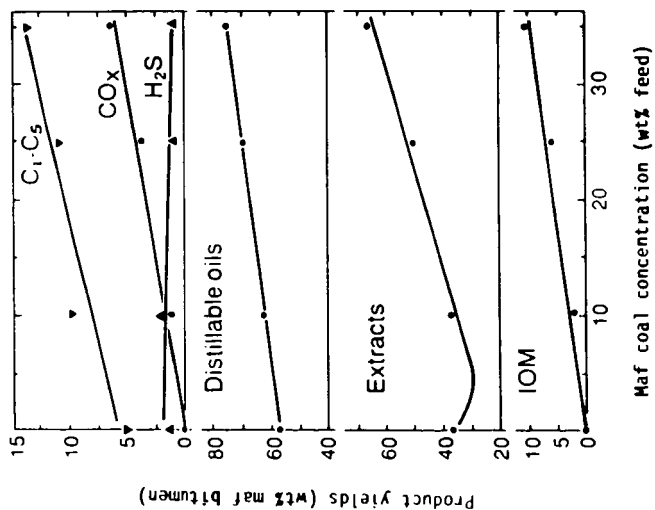


Figure 3  
Compound Class Type Distribution  
of Co-processing Product

